

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of:

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JOHN G. FRYE, JR. AND  
WAYNE R. KLIEWER

SERIAL NO: unassigned

FILED: unknown

FOR: SOLID MULTI-COMPONENT  
MEMBRANES, ELECTROCHEMICAL  
REACTOR COMPONENTS,  
ELECTROCHEMICAL REACTORS AND  
USE OF MEMBRANES, REACTOR  
COMPONENTS, AND REACTOR FOR  
OXIDATION REACTIONS

Group Art Unit:

Examiner:  
unknown

Attorney Docket  
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PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, DC 20231

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on December 6 2001

By

Carol M. Neth

(Date)

(Signature)

CAROL M. NETH

(Typed or Printed Name of Person Signing Certificate)

Sir:

The above-identified patent application is being filed concurrently with this paper and is a continuation of pending patent application Serial No. 09/491,609, filed January 26, 2000.

**5 IN THE SPECIFICATION**

Please amend the Specification of subject application.

At page 1, kindly replace the paragraph under "CROSS-REFERENCETO RELATED APPLICATIONS" as follows:

10 This is a continuation of co-pending application Serial No. 09/491,609 filed January 26, 2000, which is a continuation of application Serial No. 09/333,168 filed June 14, , 1999, now U.S. Pat. No. 6,287,432, which is a continuation of application Serial No. 08/487,945 filed June 7, 1995, now U.S. Pat. No. 6,019,885, which is a divisional of application Serial No. 08/394,925 filed  
15 February 24, 1995, now U.S. Pat. No. 5,591,315. Application Serial No. 08/394,925 is a continuation of application Serial No. 08/228,793 filed April 15, 1994, now abandoned, which was a divisional of application Serial No. 07/618,792 filed November 27, 1990, now U.S. Pat. No. 5,306,411. Application No.  
20 07/618,792 is a continuation-in-part of U.S. patent application Serial Nos. 07/457,327 filed on December 27, 1989, now abandoned, 07/457,340 filed on December 27, 1989, now abandoned, which is a continuation-in-part of U.S. patent application Serial No. 07/025,511 filed on March 13, 1987 and  
25 issued as U.S. Patent No. 4,933,054 on June 12, 1990; 07/457,384 filed on December 27, 1989; now abandoned; and 07/510,296 filed on April 16, 1990, now abandoned, which is a continuation-in-part of U.S. patent application Serial No. 07/357,317 filed on May 25, 1989, now abandoned, which are  
30 hereby fully incorporated herein by reference.

## IN THE CLAIMS

Please cancel, without prejudice, Claim 1

Please insert claims as follows:

29. A solid membrane comprising a mixed metal oxide  
5 material of a perovskite structure containing a lanthanide in  
combination with Co, Sr or an oxide thereof, provided that the solid  
membrane contains less than 13 mol percent bismuth, cerium, or  
mixture of bismuth and cerium.

30. The solid membrane of claim 29 wherein the lanthanide  
10 is lanthanum.

31. A solid multi-component membrane comprising an  
intimate, gas-impervious, mixture of a first mixed metal oxide  
material of a perovskite structure with a second mixed metal oxide  
material of a perovskite structure, wherein the first mixed metal  
15 oxide material has an electron conductivity greater than that of the  
second mixed metal oxide material and the second mixed metal  
oxide-material has an oxygen ion conductivity greater than that of  
the first mixed metal oxide material.

32. An electrochemical process for producing products  
20 which are liquid at ambient conditions from methane, natural gas or  
other light hydrocarbons which comprises

(A) providing an electrochemical cell which comprises first  
and second zones separated by a solid gas-impervious membrane  
comprising a mixed metal oxide material of a perovskite structure  
25 having electron conductivity and oxygen ion conductivity,

(B) heating the electrochemical cell to a temperature of  
from about 1000°C to about 1400°C,

(C) passing an oxygen-containing gas in contact with the membrane in the first zone, and

(D) passing methane or natural gas in contact with the membrane in the second-zone,

5 (E) recovering a substantially nitrogen-free synthesis gas comprising a mixture of hydrogen and carbon monoxide, from the second zone, and

(F) converting the recovered synthesis gas to products which are liquid at ambient conditions.

10 33. The process of claim 32 wherein (D) comprises passing a methane-steam mixture in contact with the membrane in the second zone.

15 34. The process of claim 32 wherein the membrane comprises an electron-conductive mixed metal oxide of a perovskite structure which exhibits electron-conductivity and oxygen ion-conductivity.

35. The process of claim 32 wherein air is passed in contact with the membrane in the first zone.

20 36. An electrochemical process for producing hydrogen cyanide from methane and ammonia which comprises:

25 (A) providing an electrochemical cell comprising first and second zones separated by a solid multi-component membrane comprising an intimate, gas-impervious, multi-phase mixture of an electronically-conductive phase and an oxygen ion-conductive phase,

(B) heating the electrochemical cell to a temperature of from about 1000°C to about 1400°C.,

(C) passing an oxygen-containing gas in contact with the membrane in the first zone, and

(D) passing methane and ammonia in contact with the membrane in the second zone.

5        37. The electrochemical process of claim 36 which further comprises

(E) recovering hydrogen cyanide from the second zone.

10        38. The electrochemical process of claim 36 wherein the electronically-conductive phase comprises nickel, cobalt, copper, silver, gold, platinum, palladium, rhodium, ruthenium, bismuth oxides, tin-indium oxide mixtures, praseodymium-indium oxide mixtures, cerium-lanthanum oxide mixtures, niobium-titanium oxide mixtures, or electron-conductive mixed metal oxides of a perovskite structure, or mixtures thereof.

15        39. The electrochemical process of claim 36 wherein the electronically-conductive phase comprises platinum or palladium metal.

20        40. The electrochemical process of claim 36 wherein the electronically-conductive phase comprises a praseodymium-doped indium oxide.

41. An electrochemical process for producing hydrogen cyanide from methane and ammonia which comprises

25        (A) providing an electrochemical cell which comprises first and second zones separated by a solid multi-component membrane comprising gas-impervious mixed oxide material of a perovskite structure having electron conductivity and oxygen ion conductivity,

(B) heating the electrochemical cell to a temperature of from about 1000°C to about 1400°C,

(C) passing air or oxygen in contact with the membrane in the first zone, and

5 (D) passing methane and ammonia in contact with the membrane in the second zone.

42. The process of claim 41 further comprising

(E) recovering hydrogen cyanide from the second zone.

10 43. The process of claim 41 wherein the mixed metal oxide material of a perovskite structure comprises a combination of elements selected from the group consisting of lanthanides, alkaline earth metals, Y, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr, and Nb, oxides thereof, and mixtures of these metals and metal oxides.

15 44. The process of claim 41 wherein air is passed in contact with the membrane in the first zone.

### **REMARKS**

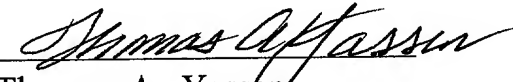
Claims 29 to 44 have been provided for examination.

20 Instant Claims 29 to 31, 33 and 35 to 42 correspond "word-for-word" with Claims 13 to 15, 67, 73 and 100 to 108 respectively, and Claims 32 and 34 are derived from Claim 66, 71 and 72 of commonly assigned application Serial Number 07/510,296 filed on April 16, 1990 which was fully incorporated by reference in the Cross-Reference to Related Applications, on  
25 page 1 of instant specification as in all intervening applications of the family. Support for Claim 32 is also found in the specification, for example at page 37, line 19 to page 38, line 30.

Applicants urge that all claims now presented, Claims 29 to 44 inclusive are in condition for allowance and request that the Examiner take such action in response to this Amendment.

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Respectfully submitted,

  
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